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Design and Control of High Temperature PEM Fuel Cell Systems using Methanol Reformers with Air or Liquid Heat Integration

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Abstract

The present work describes the ongoing development of high temperature PEM fuel cell systems fuelled by steam reformed methanol. Various fuel cell system solutions exist, they mainly differ depending on the desired fuel used. High temperature PEM (HTPEM) fuel cells offer the possibility of using liquid fuels such as methanol, due to the increased robustness of operating at higher temperatures (160-180°C). Using liquid fuels such as methanol removes the high volume demands of compressed hydrogen storages, simplifies refueling, and enables the use of existing fuel distribution systems. The liquid methanol is converted to a hydrogen rich gas with CO₂ trace amounts of CO, the increased operating temperatures allow the fuel cell to tolerate much higher CO concentrations than Nafion-based membranes. The increased tolerance to CO also enables the use of reformer systems with less hydrogen cleaning steps and requirements for hydrogen purity, reducing the complexity of the reformer systems. Using hydrogen containing CO, affects the steady-state as well as dynamic electrical performance of the fuel cell, but stable operation is still possible with concentrations up to 3%. The typical polymer used in HTPEM fuel cells is polybenzimidazole (PBI), doped with phosphoric acid for proton conduction. The work will present a few different methanol reformer concepts, some experimental results of details related to reformer gas quality, control of burner temperature and the aspects of implementing advanced modeling based control approaches using the commercial Serenergy H3-350 methanol reformer system as an example.

1 Introduction

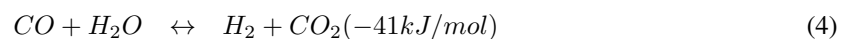
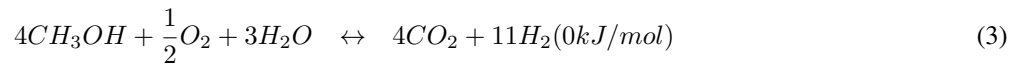
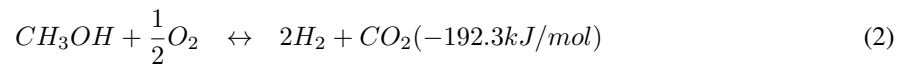
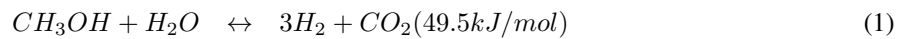
Fuel cells are an efficient means of converting the chemical energy in a fuel directly into electricity. Different type of fuel cells exist characterized by different electrolytes, operating temperatures, pressures and preferred choice of fuel. They have shown promising performance and results used within forklifts and material handlers [1, 2, 3, 4, 5], automotive and hybrid electrical systems [6, 7, 8, 9, 10, 11, 12], auxiliary and uninterruptible power systems [13, 14, 15, 16, 17, 18, 19].

Polymer electrolyte membrane fuel cells are some of the most commercially advanced fuel cell technologies. In the recent years high temperature PEM (HTPEM) fuel cells have received increased attention [20, 21] due to their robust performance which mainly is the results of operating at higher temperatures; typically 160-180°C [22, 23, 24, 25]. Low temperature PEM fuel cells typically use polymer membranes made by sulphonated polytetrafluoroethylene (PTFE) for ionic conduction (mainly Nafion). The polymer relies on the presence of liquid water for proton conduction, which limits its operating conditions to temperatures below 100 °C. These types of PEM fuel cells are among the most efficient on the market. The presence of liquid water is however not without challenges, the amount of liquid water in membrane electrode assembly (MEA) is of high importance and small changes in humidity and temperatures of the inlet gasses can have severe impact on performance. High humidity can often resulting droplet formation, which in turn blocks flow channels of components. Very hot and drying conditions can complete dehydrate the fuel cell and leave it non-operational.

The higher operating temperatures introduce the need and possibilities of using high temperature PEM fuel cells, using different polymer membranes. Polybenzimidazole (PBI) based MEAs are of particular interest, and they use phosphoric acid as an ionic conductor removing issues of water management and humidification challenges. However moving to higher temperatures is not without problems, the materials (membranes, catalysts, stack components) are less developed, starting time is longer and performance is lower. But systems can often attain comparably higher efficiencies due to the ease of cooling, reduced requirements to inlet fuel quality and possibility of using non-hydrogen based liquid fuels such as methanol. Adding a fuel reformer introduces additional complexity to the system that requires proper control strategies in order to obtain a reliable and efficient system performance. This work will present some of the challenges and strategies involved with such system design.

2 Methanol reforming

Using a liquid fuel, such as methanol for fuelling a fuel cell system removes some of the challenges involved with hydrogen-based fuel cells, such as fuel availability and handling, investments in infrastructure and on the volumetric challenges in storing hydrogen. Methanol is the simplest alcohol, it is a commonly used component in the industry which is widely available. The temperatures of the reforming process are quite low (220-300°C) compared to the reforming of other commonly known fuels, such as natural gas or diesel (>700°C). Low cost CuZn-based catalysts can be used, and different operating methods are possible. In equation 2 the most common reforming reactions are listed.



The most common efficient reforming method is the steam reforming reaction in equation 2 which is an endothermic reaction requiring an addition of heat to the process. The other reactions (equation 3 and 4) adds air to the reforming process, which in turn partially combusts part of the developed gasses. In the autothermal reaction equation 4, the heat generated by oxidation of the fuel is balanced to match the heat requirement for reforming. The resulting gasses of the steam reforming reaction are H_2, CO_2, H_2O, CO and unconverted methanol. Related to the fuel cell stack, it is important to minimize the different pollutants in the hydrogen rich reformer output gas, such as CO and residual methanol, which will effect both the immediate stack performance and also lifetime [26, 27, 28]. Furthermore the water-gas-shift reaction (equation 4) is an important process that typically occurs parallel to the steam reforming reaction removing CO.

In order for methanol reformer system to yield optimal efficiencies, proper heat integration is needed, i.e. all practically usable waste heat should be utilized. Different major heat consumers exist in high temperature PEM fuel cell systems, such as fuel evaporation and reformer reaction heat requirements. And the heat sources available in the following shown cases are fuel cell stack waste heat and anode exhaust hydrogen. Different system configurations are possible

for transferring heat from and to these sources, the following examples will present a system topology using air as the main heat carrier for transferring heat in the system, mainly heat from the fuel cell stack to facilitate the evaporation of the fuel. A second system is presented using a heat transfer oil as medium for transferring heat in the system using two circuits.

2.1 Air cooling system configuration

Multiple authors have presented work on using air as the cooling medium in HTPEM fuel cell stacks, either as cathode air cooled stacks, where cathode air at very high stoichiometries is used for controlling the temperature of a fuel cell stack, not requiring additional cooling channels in the construction of the fuel cell stack. This often yields very simple systems, because fewer Balance-of-Plant components are needed and stack design often is simpler. The drawback is the additional optimization of the stack flow fields and manifolds to ensure low pressure drops under such increased flow conditions, in order to use low power consuming air supplies. Obtaining a uniform cell temperature is another challenge. In other cases separate cooling circuits are present, and the cooling and temperature control can be done separately using an additional air cooling system. In the case where a methanol reformer system is introduced, the waste heat from the fuel cell stack can effectively be used as process heat for evaporating the methanol/water mixture used by the system. Figure 1 shows a typical schematic of a reformed methanol HTPEM fuel cell system using air based cooling and heat transfer. The heat required by the methanol reforming process, which normally takes place at temperatures higher than the 160-180°C of the fuel cell stack, can be utilized from the excess hydrogen exiting the fuel cell stack. An excess amount of hydrogen is normally supplied to the fuel cell stack, i.e. the fuel cell stack is running at a stoichiometry of $\lambda = 1.2 - 1.3$ in order to avoid dilution effects and ensure proper performance, because of the presence of residual gasses such as CO_2 , CO , H_2O and CH_3OH . This anode waste gas is catalytically combusted in a burner, and the heat is used to keep the reformer active at the desired operating temperature of around 280°C. Air is supplied to the anode waste gas, which also supplies the oxidant for the combustion process, and the air flow is adjusted to decrease the resulting flue gas temperature, which is sent across a network of heat exchangers that pre-heat incoming gasses and supplies the catalyst bed with heat.

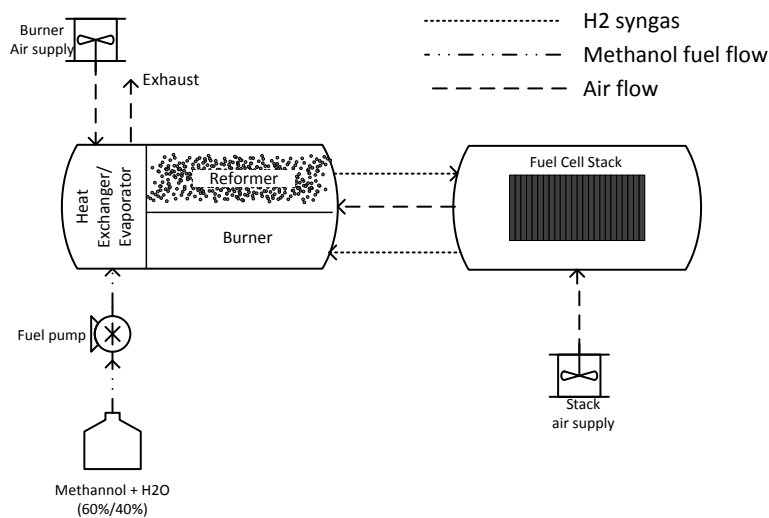


Figure 1: Schematic of a methanol reformer system using air for heat transfer.

An example of a commercial HTPEM fuel cell system as the one exemplified above, is the Serenergy H3-350, which uses a similar a operating principle as shown in figure 1. The system can be seen in figure 2, and can, as an example, be used as an off grid battery charger for 24V batteries, as it has a DC/DC controlled power output able to charge for example a 24V lead-acid battery packs.



Figure 2: The Serenergy H3-350, 24V off grid battery charger.

The system includes the integrated fuel cell stack and methanol reformer system, DC/DC converter and Balance-of-Plant components and a small fuel buffer tank, and requires a fuel tank dimensioned to the desired runtime of the system. During start-up, such systems as this can often be heated either by electrical heating elements because of the low mass and fast temperature transients, but also using the available methanol fuel for heating a system is possible from low temperatures and yields higher round-trip efficiencies.

2.2 Liquid cooling system configuration

When increasing power levels, losses also increase and in order to keep system size compact and efficiency high, liquid heat transfer is often an advantage, an example of such a system configuration is shown in figure 3.

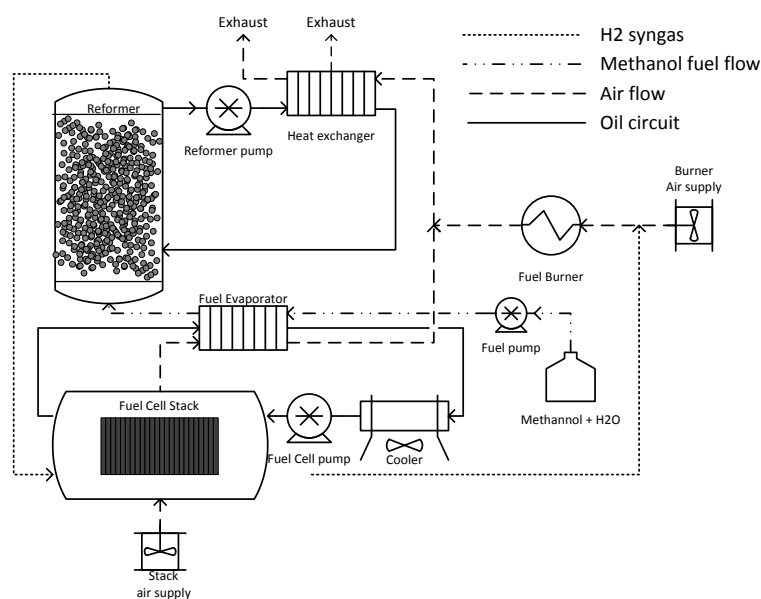


Figure 3: Schematic of a methanol reformer system using a liquid heat transfer medium.

Using a liquid cooling media requires additional cooling channels in the fuel cell stack and additional components for circulation and heat exchange. The advantages are often that more industrial off-the shelf components are available and not low pressure specialized compressors and blowers requiring optimized manifolds and custom made heat integration. Because circulation of liquids often can be increased efficiently compared to air compressors and fans, more uniform temperature profiles can be expected on both reformer and fuel cell stack.



Figure 4: The Serenergy H3-15k system with an integrated methanol reformer and liquid cooled HTPEM fuel cell stack.

2.3 Discussion

Many challenges exist when introducing reformers in fuel cell systems, the advantages are related mainly to the added fuel flexibility, not relying on a pure hydrogen source from either a compressed storage, metal hydride or liquid hydrogen supply. The following topics will shortly be discussed in order to illustrate the possibilities of reformed methanol fuel cell systems using HTPEM fuel cells:

- System start-up time
- Aspects of added system complexity and system control and load following capabilities

2.4 System start-up

Start-up times of HTPEM fuel cell systems, are of varying importance, the main importance is the balance of fuel cell and battery energy and the average power cycle demand. Systems where the fuel cell system composes the main part of the power available need fast start up, and often also power delivering capabilities. Systems that act as range or run time extenders in automotive vehicles or auxiliary power units, are often not as time critical. HTPEM systems require certain minimum temperatures in order to operate reliably and efficiently and often use electrical heating. The disadvantage is that the system round trip efficiency is diminished because they need to operate for a longer period of time in order to re-produce the energy needed for start-up. Several authors have examined different ways of heating HTPEM fuel cell stacks, and when it comes to an efficient way of handling this disadvantage of using electrical energy for heating, using the reformer fuel directly in catalytic combustion can be used.

Figure 5 shows the temperature profile of a heat exchanger reformer (similar to the one described in [29]) when starting up on a liquid methanol reformer fuel, only requiring a small amount of electrical heating for the initial evaporation of

the methanol/water mixture. A better round trip system efficiency is gained because the heat needed for system heating is more efficiently used.

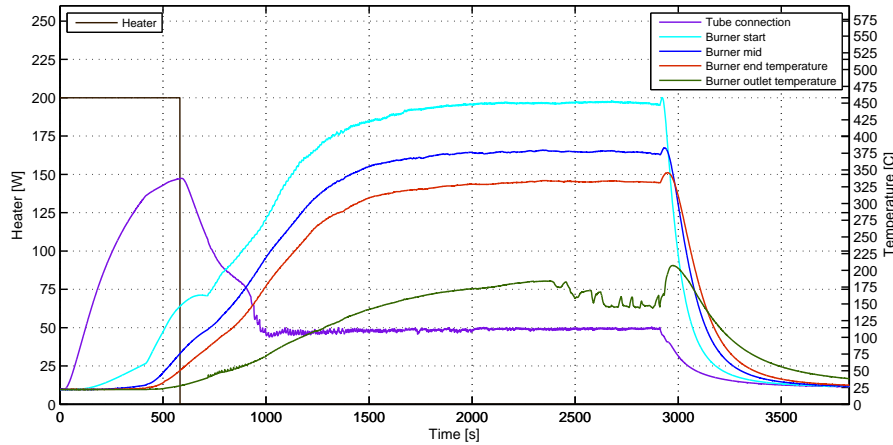


Figure 5: Example of reformer start-up using catalytic combustion of a 60/40 methanol water mixture.

2.5 Added system complexity and system control strategies

When reformers are added to fuel cell systems, several other types of fuels than pure hydrogen become possible, including liquid fuels, such as methanol. Adding a reformer system often increases the complexity of the fuel cell system and can add restrictions to the output performance. In order to handle this added complexity, the development of proper control strategies is relevant. Several possibilities exist for ensuring safe performance and reliable and efficient operation. Besides safety, i.e. keeping the system within certain operating temperatures, gas compositions and efficiency; many other states are also of critical importance when it comes to system lifetime. For HTPEM fuel cell systems it is of key importance to avoid anode starvation [30, 31, 32, 33] because it increases anode overpotentials and rapidly accelerates the fuel cell degradation due to carbon corrosion, because of the elevated operating temperatures. Methanol reformer systems are more prone to this type of degradation because of the additional delay in gas transport and chemical conversion. In order to handle a change in load current I , the fuel flow \dot{m}_{Fuel} must be changed. Such a change requires initially an increase in the evaporation of the additional water/methanol mixture flow and additional reformer conversion, before the resulting in the increased hydrogen flow into the fuel cell stack. Depending on system size and configuration the time delay involved can be significant, and to further complicate things, temperature transients are introduced in all system components, evaporator, reformer, burner and fuel cell stack. In the shown air cooled system on figure 1, the fuel cell stack cooling is controlled by a separate actuator, an air blower, but initially the evaporator will require an increased heat input which will not be available before the load change has occurred. When an evaporator operates of the waste heat of the fuel cell, the temperature is usually far higher than the boiling point of the mixture, but efficient heat transfer should be ensured. The methanol reformer in the air cooled system could also encounter problems because the anode exhaust hydrogen also is delayed, which affects the burners ability of adding increased amounts of heat to the endothermic reformer reactions. These transport delays often change the temperature of the system components during load changes, and reformer reactions and fuel cell stack operation is highly dependent on temperatures and the gas composition of the

reformat gas. One solution to this problem is power conditioning, i.e. constraining the rate at which the current on the fuel cell stack changes, either by a DC/DC converter or on the application using the fuel cell power. Other solutions require detailed knowledge of the system thermal dynamics, residence time in the reformer and how these variable conditions affect the gas composition and hydrogen flow into the fuel cell stack.

The prediction of the needed pump flow for a certain fuel cell current is an important parameter, especially if high efficiencies are required, as the system efficiency η_{system} is strongly dependent of the pump flow of fuel \dot{m}_{fuel} in the system:

$$\eta_{system} = \frac{P_{output}}{P_{input}} = \frac{U_{FC} \cdot I_{FC} - P_{BOP}}{\dot{m} \cdot LHV_{CH_3OH}} \quad (5)$$

The system efficiency depends on the fuel cell stack performance, dictated by the fuel cell voltage U_{FC} and current I_{FC} and the balance-of-plant power consumption P_{BOP} . In order to choose the correct fuel flow desired for a certain load current and anode stoichiometry, detailed knowledge of the methanol reformer temperature and other system variables are required. Figure 6 shows an example of a feed forward control strategy for a methanol reformer system using air cooling.

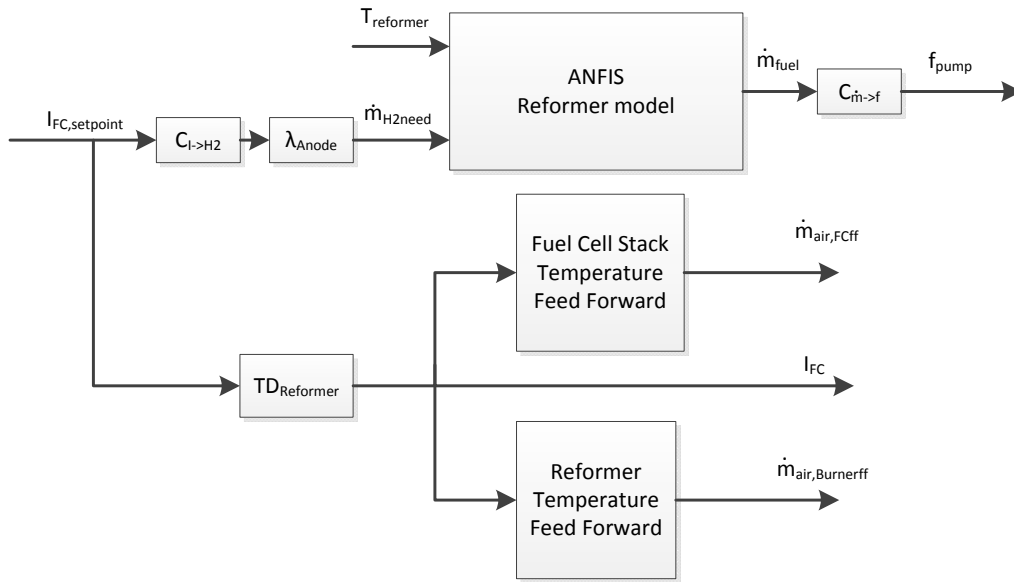


Figure 6: Feedforward part of a methanol reformer system control strategy using ANFIS modelling.

The fuel pumping frequency f_{fuel} , air mass flow for the fuel cell stack and burner and the delayed fuel cell current due to system delay all are predicted using simple models of the system and an Adaptive Neuro-Fuzzy Inference System (ANFIS) model for the prediction of the reformer output gas composition depending on temperature $T_{reformer}$ and desired hydrogen mass flow $\dot{m}_{H_2 need}$. Combining such a feed forward control strategy with a properly designed control of the different temperatures and other critical states of the system will allow high efficiency and the possibility of controlling certain internal states of the system not normally measurable, such as CO gas concentration and anode stoichiometry λ_{H_2} . Introducing more adaptive control strategies and detailed information of the degradation performance of the system could also further improve such models and control strategies to ensure improved lifetime and good performance over the entire lifetime of the fuel cell system.

System efficiencies obtainable by reformed methanol fuel cell systems using HTPEM fuel cells can be increased by proper heat integration and attention to control of critical system states. In order to achieve the highest possible system efficiencies, improvements are needed of fuel cell catalysts, to tolerate even higher CO concentrations without performance losses. Furthermore if reformer catalysts are improved to good performance at reduced Steam-to-Carbon ratios, lower system heat requirements are needed for fuel evaporation. Finally improving reformer catalyst activity at temperature closer to the fuel cell operating temperature will ensure better utilization of waste heat and possibly lower equilibrium CO in the reformat.

3 Conclusions

This work has presented different important aspects to take into consideration when using methanol reformer system in combination with high temperature PEM fuel cell systems. The introduction of methanol reformers complicate system performance by introducing additional system components and processes. But with properly design control strategies the system can be brought to efficient and reliable performance. The important states to control and evaluate in HTPEM fuel cell methanol reformer systems are: reformer output CO concentration, presence of unconverted fuel, hydrogen stoichiometry in the fuel cell stack and fuel dynamics from load change to available fuel cell stack output power without compromising the lifetime of the fuel cell stack.

4 Acknowledgements

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